DESIGN AND CONSTRUCTION OF AN ULTRA-LOW-BACKGROUND 14 GERMANIUM CRYSTAL ARRAY FOR HIGH EFFICIENCY AND COINCIDENCE MEASUREMENTS

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ABSTRACT

Physics experiments, environmental surveillance, and treaty verification techniques continue to require increased sensitivity for detecting and quantifying radionuclides of interest. This can be done by detecting a greater fraction of gamma emissions from a sample (higher detection efficiency) and reducing instrument backgrounds. A current effort for increased sensitivity in high resolution gamma spectroscopy will produce an intrinsic high-purity germanium (HPGe) array designed for high detection efficiency, ultra-low-background performance, and useful coincidence efficiencies. The system design is optimized to accommodate filter paper samples, e.g., samples collected by the Radionuclide Aerosol Sampler/Analyzer (RASA). The system will provide high sensitivity for weak collections on atmospheric filter samples, as well as offering the potential to gather additional information from more active filters using gamma cascade coincidence detection.

The current effort is constructing an ultra-low-background HPGe crystal array consisting of two vacuum cryostats, each housing a hexagonal array of 7 crystals on the order of 70% relative efficiency each. Traditional methods for constructing ultra-low-background detectors are used, including the use of materials known to be low in radioactive contaminants, the use of ultra pure reagents, clean room assembly, etc. The cryostat will be constructed mainly from copper electroformed into near-final geometry at Pacific Northwest National Laboratory (PNN)L. Details of the detector design, simulation of efficiency and coincidence performance, HPGe crystal testing, and progress on cryostat construction are presented.

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OBJECTIVES

This research will produce a high efficiency array of germanium crystals in an ultra-low-background system to provide improved gamma spectroscopy sensitivity. The system will be composed of two separate copper cryostats, each housing arrays of 7 HPGe crystals to provide high detection efficiency for large sized samples (e.g., filter paper media). The high detection efficiency serves to increase source counts for a given measurement, improving the high-leverage term in the S^2/B figure of merit (FOM). The approach also combines the inherent excellent energy resolution of intrinsic germanium with ultra-low background detector construction and operation techniques to drive down the background term. For appropriate isotopes and measurement scenarios, additional sensitivity gains will be realized through the use of gamma-gamma coincidence techniques.

Measurements of atmospheric particulates collected on filter papers are one of the principal sample analyses that this effort is pursuing. The intention is to provide the highest sensitivity gamma spectroscopy measurement possible prior to radiochemical or other destructive processing of a sample. Filter papers collected by automated ground-based particulate samplers (e.g., RASA) or aircraft-based collectors are examples of potential samples. High interest samples (e.g., where other techniques have provided a tip-off) with initial negative results from field-based measurements are prime candidates for analysis. While a primary focus is currently on intact filter paper samples, the system will also provide an excellent measurement capability for chemically purified samples, and also has the potential to provide an outstanding gamma-gamma coincidence analysis of low-dose Neutron Activation Analysis samples.

Design Philosophy

This system is designed to make the best possible use of the relatively small number of radioactive decays encountered in measuring a weak sample. This requires reducing backgrounds to the greatest extent possible so that sample activity is the dominant signal in the detector. Also required are the highest possible gamma detection efficiencies, meaning more of the emitted gammas are seen as signals in the detector. Finally, some isotopes have very specific nuclear decay signatures, and coincident gamma emissions, which can improve selectivity for these isotopes in a coincidence-capable detector.

Used all together, these methods can greatly enhance the sensitivity of a gamma spectroscopy system. Translated into design rules, they become three main focus areas for the instrument: ultra-pure materials, highly effective and low-background shielding, and array design to maximize the detection efficiency for both single and coincident gammas.

RESEARCH ACCOMPLISHED

Significant progress has been accomplished toward construction of the first 7-crystal cryostat. The cryostat has been designed and extensive Monte Carlo modeling of the radiation detection performance is complete. New copper electroforming capability has been established, and parts for the first cryostat have been electroformed and machined into final form. Princeton Gamma Tech (PGT) has completed refurbishing eight existing PNNL HPGe crystals for this effort, and these crystals are being mounted and tested in a new mount design. A new revision of PNNL-developed low-background front-end electronics has been designed for this work and is being manufactured (Aalseth et al., 2004). This new instrument also represents significant complexity in data acquisition and analysis, and work proceeds to develop these necessary tools as well. Currently, the research team is working primarily on cryostat construction, vacuum and thermal testing. After successful testing, the first cryostat will be populated with HPGe crystals.

System Design

The current system design was developed following PNNL's experience with the Multi-Element Gamma Array (MEGA) project (Kazkaz et al., 2003). The essential feature is to construct two HPGe 7-crystal arrays facing one another. In each array (see Figure 1), the crystals will be housed in a single cryostat / vacuum enclosure. Each crystal is mounted in an individual mounting package; this will reduce direct handling of the crystals, and allows relatively straightforward removal of a crystal for repair. The design of the crystal-mounting package draws on a number of recent designs in the double beta decay arena, and attempts to minimize materials between the crystals for improved detection of Compton scattered gamma-rays. Housing the array in a single enclosure also allows closer placement of the crystals, improving detection efficiency for the system. These two aspects, improved Compton-

scatter detection and better solid-angle coverage, are significant technical advantages over current cryostat designs, and make important contributions to the expected performance of the system. One drawback of the design is that failure of a single crystal requires the entire 7-crystal unit to be warmed and opened for repairs. The technical performance advantages of an array of crystals in a single enclosure over deploying crystals in separate cryostats far outweigh the increased risks when service is needed. Typical cryostats can go for many years without requiring such service.

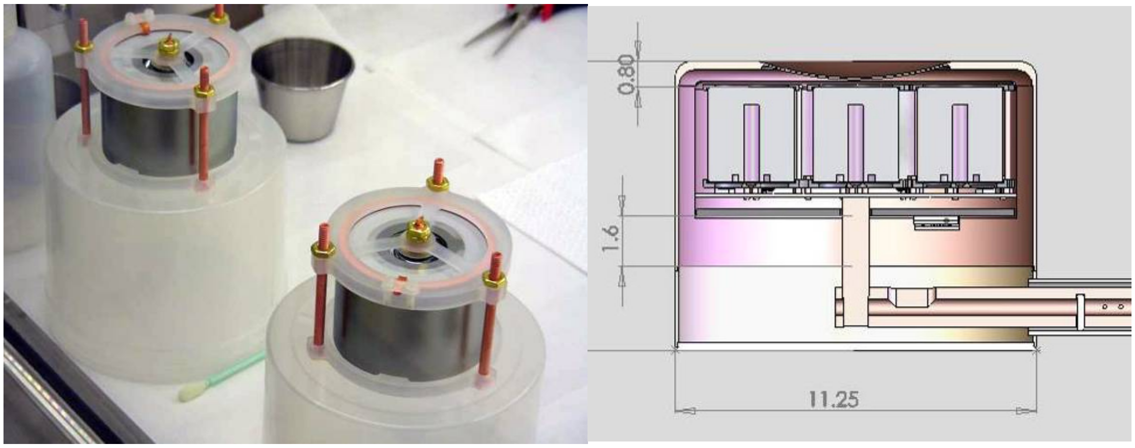

Figure 1. The HPGe crystals are housed in individual crystal mounting packages that can be removed from the cold plate if repairs are necessary (left). The 7 crystals in their individual mounts will be bolted to the cold plate, and enclosed by a single copper IR shield. The system will also include an intermediate "floating" shield to enhance thermal performance. The design includes a thin copper entrance window to improve detection efficiency for lower energies (dished portion at the top of the figure on right).

The two detector arrays will be housed in a lead shield depicted in Figure 2. The shield includes 16" of lead shielding, radon purge using boil-off nitrogen from the liquid nitrogen Dewar that cools the HPGe cryostats, and cadmium sheets exterior to the lead to reduce neutron events. The inner 2 layers of lead are low background lead or ultra-high-purity copper, depending on the location (depth) of the deployed system. An electric motor is used to roll the lead door out of the shield, allowing easy access for sample placement and detector maintenance.

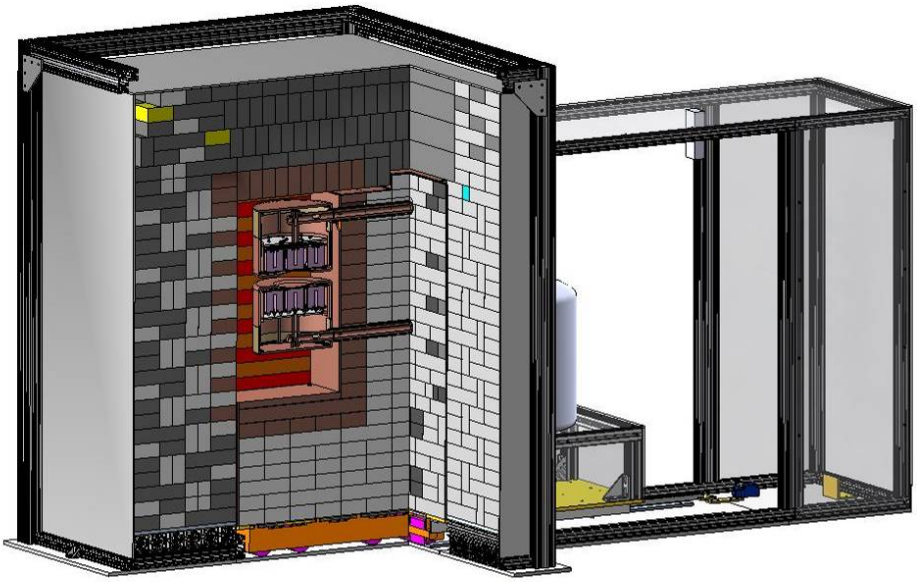

Figure 2. Depiction of the two 7-crystal arrays in a 16" thick lead shield. The door of the shield opens to allow sample changes, as well as access for maintenance without un-stacking the lead.

Performance Modeling

Simulations of the system detection efficiency performance and measurement of mixed fission products were conducted using the GEANT4 (G4) framework (Agostinelli 2003, Allison 2006). A diagram of the detector geometry is shown in Figure 3. The simulated geometry consists of two arrays of seven large p-type HPGe crystals looking head-on. Each crystal is 62 mm in diameter, 70 mm long, and has a dead layer of 0.25 mm. The actual dead-layer thickness of the crystals used will likely be a bit larger, reducing lowest-energy performance slightly. Each crystal array is surrounded by a 0.1-mm-thick IR shield, and a 5-mm-thick vacuum chamber wall with a 1 mm thick entrance window. A BGO Compton suppression detector was placed in the simulation surrounding the two crystal arrays for analysis of the potential benefit of such a future addition to the system. Decay particles were generated in a 100 mm square (about the size of a folded filter sample) 26 mm from the front of the crystals.

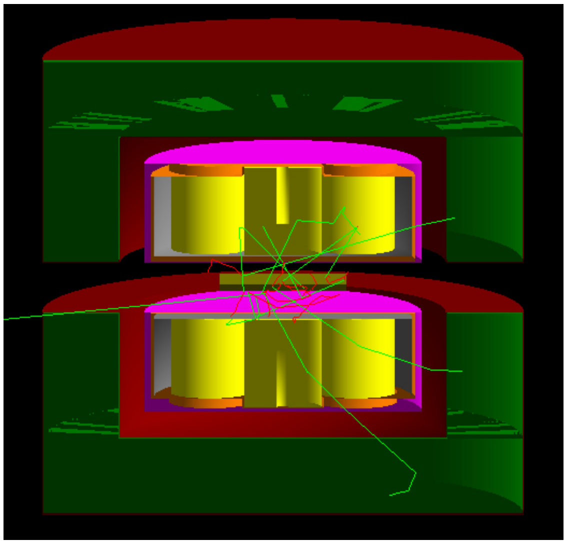

Figure 3. The GEANT-4 geometry used for simulation of the 14-crystal array.

Radioactive decay products for each isotope were generated based on the PNNL Coincidence Lookup Library (CLL) (Warren 2006). The CLL provides the cascade information for each isotope, such as probability of cascade and particle energies involved, based nuclear data from a version of the ENSDF database. The simulation package uses this cascade information to generate the decay particles for each isotope, including the generation of coincident gamma cascades.

The activity for each isotope was determined from a published series of ORIGEN2 calculations (Perkins 1997) of the time dependant fission product inventory subsequent to an HEU based nuclear explosion. The published tables were used as a starting point for a separate ORIGEN2 calculation that determined the average activity of each isotope from five to six days post-event. A sample size of 10^9 fissions and a measurement period of 24 hours were assumed. A total of 80 fission products and 40 million decays were simulated in order to get a fair representation of the mixed fission product spectrum likely to be present.

After the number of decays for each isotope was determined, they were simulated independently through the detector model. The individual energy spectra collected were then summed to produce the mixed fission product spectrum. Isotopic-specific weighting was applied to a few specific isotopes for the summing process. For most isotopes, the weighting was 1.0. For the noble gas isotopes, the weighting was zero and for the iodine isotopes it was 0.333, except for I-132 which was set to 1.0. This weighting factor allowed the incorporation of the expected reduction in signals for the noble gases and most of the iodine isotopes due to chemical fractionation prior to collection by a particulate sampler. These singles histograms were analyzed using Genie (Canberra) and the coincidence histograms were analyzed using the PNNL-developed Multiple Isotope Coincidence Analysis (MICA) code (Warren et al., 2006).

Analysis of simulation results indicates that the RN Labs instrument should achieve roughly an order of magnitude improvement in sensitivity over measurement with a single ultra-low background crystal (during a 24-hour count). Table 1 presents the determination limit, L_Q, required to achieve a 10% result in a 24 hour measurement from day 5 to day 6, after an event. The table includes several high-yield fission products. In this analysis, the background from one of the “twin” detectors (Brodzinski et al., 1990) operating at ~ 1000 meters water equivalent (mwe) was used as an estimate for the background of each individual crystal in the 14-crystal array. The simulation results for one central crystal were used to calculate the single crystal value.

Table 1. **Determination limit comparison and estimated detection limit.**

		L_Q for 10% (fissions)			L_D (fissions)
Isotope	**Energy (keV)**	**Single Crystal**	**14 Crystal Array**	**Analysis Method**	
^{95}Zr	756	1.6E07	1.4E06	Super Sum	1.4E05
^{99}Mo	739.5	1.2E07	1.7E06	Summed Spectra	1.9E05
^{103}Ru	497	8.9E06	9.2E05	Super Sum	1.4E05
^{131}I	364*	2.3E06	2.8E05	Super Sum	5.0E04
^{132}Te	228*	9.0E05	1.4E05	Super Sum	3.7E04
^{140}Ba	537	7.9E06	7.5E05	Super Sum	9.6E04
^{140}La	1596	4.7E06	6.8E05	Super Sum	4.1E04
^{141}Ce	145*	3.8E06	7.9E05	Summed Spectra	1.7E05
^{143}Ce	293*	3.5E06	5.0E05	Super Sum	9.6E04
* Potential background continuum contributed by atmospheric ^{7}Be has not been taken into account.					

Table 2 presents a comparison of the projected system detection limits (from the far right column of Table 1) with a notional single crystal with a typical above-ground shielded background. For this comparison, the background from a commercial low-background detector installed in typical shielding at PNNL was used. The system does not have active anti-cosmic shielding or radon exclusion. Figure 4 shows a comparison of the surface detector background and postulated 14-crystal background at ~1,000 mwe. Detection efficiencies are based on the same single crystal simulation results for a central crystal of the RN Labs array.

Table 2. **Detection limits for high fission yield gamma emitters single crystal at surface vs. 14-crystal array at 1000 mwe.**

		L_D (fissions)		**Improvement Factor**
Isotope	Energy (keV)	Single Crystal	RN Labs	
^{95}Zr	756	9.4E06	1.4E05	67
^{99}Mo	739.5	7.0E06	1.9E05	37
^{103}Ru	497	6.9E06	1.4E05	49
^{131}I	364	2.2E06	5.0E04	44
^{132}Te	228	9.8E05	3.7E04	26
^{140}Ba	537	5.6E06	9.6E04	58
^{140}La	1596	1.3E06	4.1E04	32
^{141}Ce	145	4.2E06	1.7E05	25
^{143}Ce	293	3.5E06	9.6E04	36

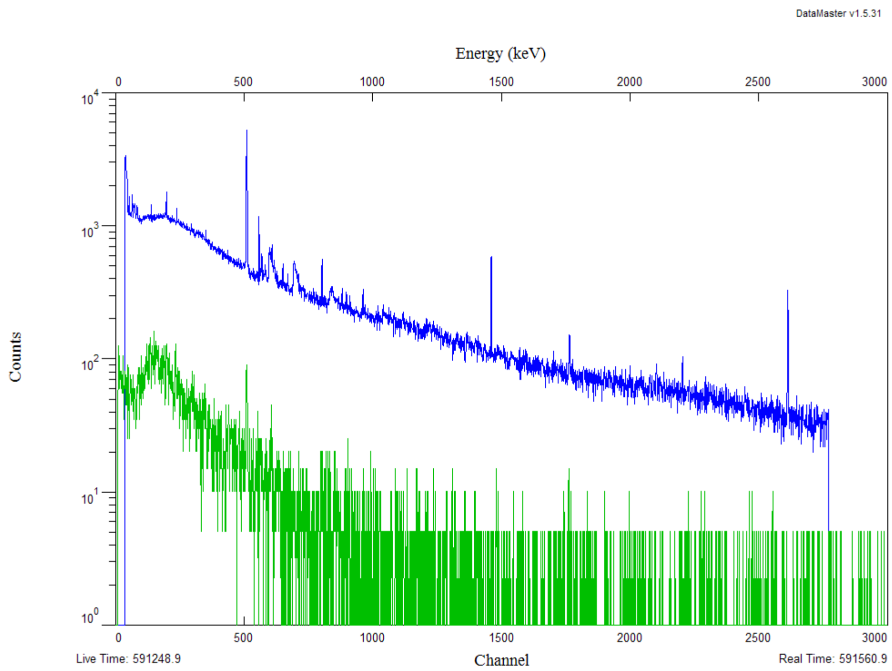

Figure 4. A comparison between the background of a single shielded commercial low background detector, and potential background performance of this 14-crystal array.

At this stage of our analysis, a few fission product isotopes have been identified that are detected with better sensitivity in coincidence mode than in singles mode by the 14-crystal instrument. Coincidence signatures for the same MFP measurement scenario used for singles mode analysis (24-hour count from day 5 to day 6) were investigated. In many cases, sensitivity gains realized by background reduction do not overcome the efficiency loss associated with detecting the coincident signature. This situation is exacerbated by the restricted count length—for isotopes with appropriate half-lives, longer count lengths would allow useful collection of data at very low background rates. Longer count times allow the sensitivity of the coincidence mode to overtake that of singles mode for additional mid- to long-lived isotopes.

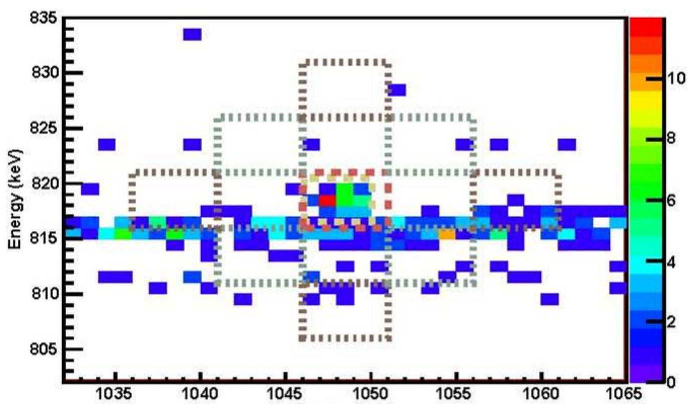

Figure 5. Simulation results for detection of ^{136}Cs using the coincident 815.5 and 1048 keV gamma-rays. This peak lies next to the Compton scatter ridge formed by full-energy deposition of the ^{140}La 815 keV gamma, and Compton scatter of a coincident, higher energy ^{140}La gamma (e.g., 1,596 keV). This plot is from a simulation of 10^9 fissions.

Figure 5 shows the 2-D coincidence plane for analysis of ^{136}Cs. This figure represents simulation of 10^9 fissions, resulting in a more sensitive determination of ^{136}Cs than achieved with the singles analysis. In this case, the ridge running across the figure at 815 keV is associated with ^{140}La, while the peak at the center is made up of ^{136}Cs 818.5 – 1048 keV coincidence counts.

Europium-156 is another isotope that can be detected with higher sensitivity using coincidence techniques. In this case, the most intense gamma is emitted at 811.8 keV. As with ^{136}Cs, this line is obscured in singles mode due to the strong ^{140}La line at 815 keV. The 10^9 fissions coincidence detection plane for the 646 and 1231 keV coincidence signature is shown in Figure 6. While this

signature isn't as dramatic the one in Figure 5, it represents a positive detect of ^{156}Eu in the simulated data at near detection limits. Europium-156 is not detected in singles mode; in fact, the ^{156}Eu activity would need to be elevated by roughly a factor of three above the simulated value to exceed the detection limit in singles mode.

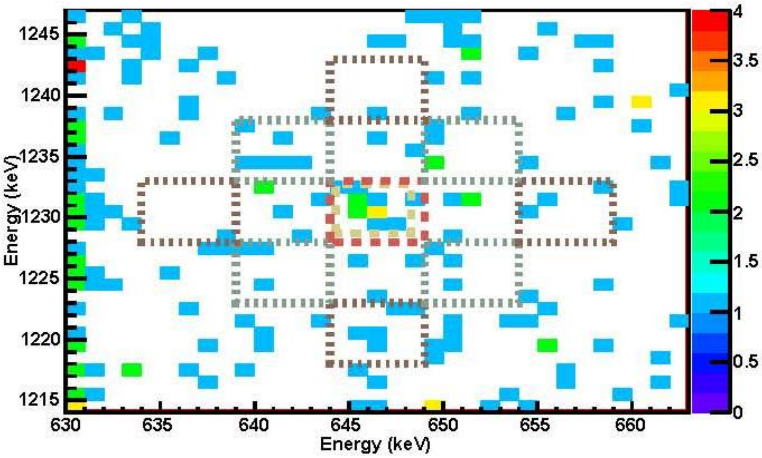

Figure 6. Simulation results for detection of ^{156}Eu using the coincident 646 and 1,231 keV gamma-rays. The isotope is detected slightly above the detection limit. This plot is from simulation of 10^9 fissions.

Copper Electroforming and Assay

For over 20 years, PNNL has made improvements to the radio-purity and mechanical properties of electroformed copper. Recent inductively coupled plasma mass spectrometry analyses of this electroformed material have provided the lowest measured values of thorium impurities in copper. These techniques are being used to produce ultra-low-background cryostat parts for the current effort. Although copper electroforming is a long-standing capability at PNNL, electroforming baths with sufficient purity and capacity for the large parts required for this effort were not available at the beginning of the project. Two new large baths have been established and are working at full capacity to produce cryostat parts. These baths are capable of producing parts like pieces for the main body of the cryostat (~ 11" diameter), as well as long parts like the cross-arm and cold finger (~30" long). Figure 7 shows the cold finger for the first cryostat while it was being electroformed, as well as one of the large diameter parts for the cryostat.

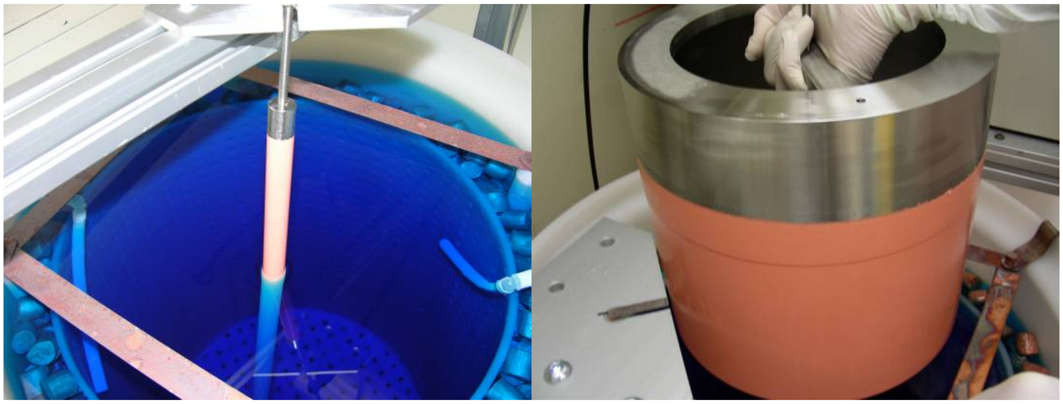

Figure 7. Electroforming the cold finger and one of the large-diameter parts for the first cryostat. These photographs are from early in the electroforming process—both parts have since been completed and sent on for machining.

HPGe Crystals

Eight HPGe crystals have been received from PGT and have passed acceptance testing. These crystals were available at PNNL from prior projects, and were refurbished by PGT for this research. After acceptance testing, the crystals were removed from the delivery cryostat and kept in clean room conditions in a dry nitrogen storage cabinet. Storage periods ranged from a few weeks to several months. The crystals are being remounted into crystal packages shown in Figure 1, in preparation for populating the first 7-crystal array. A purchase order for additional new crystals for the 2nd cryostat has been placed with Canberra. The new crystal mounting technique has been successfully demonstrated. One of the main drawbacks to this method is the poor thermal coupling between the cold mass of the cryostat and the HPGe crystal. Typical single-crystal commercial cryostats will cool the crystal in less than 6 hours, while thermal equilibrium is not reached for ~48 hours in this new mount. While this is inconvenient in the event of repair work, thermal cycling of ultra-low-background systems is avoided as much as possible. Thus, this long cool-down period should not be a significant inconvenience for this system.

Analog and Front-end Electronics

Semiconductor radiation detectors like HPGe produce signals too small to be observed without amplification. Typically, the first stage of amplification is placed only a few cm from the crystal, and is called the “Front-End” of the signal amplification and conditioning chain. Because it is close to the crystal, the radiopurity of this front-end is important. Because it contains active and passive electronic components, it is more challenging to purify of radioactive backgrounds than a bulk material like the copper used for the cryostat. Prior work has addressed this challenge by developing first a hand-assembled Low-background Front-End Package (LFEP), a second LFEP design that was suitable for production with standard electronics industry methods, and finally, a significantly smaller LFEP design that is to be used in this system. The evolution from a commercial front-end through the three LFEP designs can be seen top left-to-bottom right in Figure 8.

The LFEP works with the rest of the HPGe preamplifier to form a charge-integrating loop, converting the charge deposited in the HPGe by ionizing radiation into a voltage change that can be accurately measured by the digitizers described in the next section. For this work, custom 4-channel charge-integrating preamplifier boards have been prepared by Bridgeport Instruments to match the electrical characteristics of the LFEP units.

Data Acquisition and Analysis

Data will be acquired from the 14 crystals and anti-cosmic shielding using X-Ray Instrumentation Associates (XIA) Pixie-4 cards, which provide four channels of digitization using 14-bit ADCs sampling at 75 MHz. This will allow digitization of event pulses received from all 14 crystals, if desired. Recording the pulse waveforms allows further background reduction through rejection of spurious pulses. This hardware platform offers ~13 ns time-stamps on all events and thus allows accurate reconstruction of coincident interactions in multiple crystals.

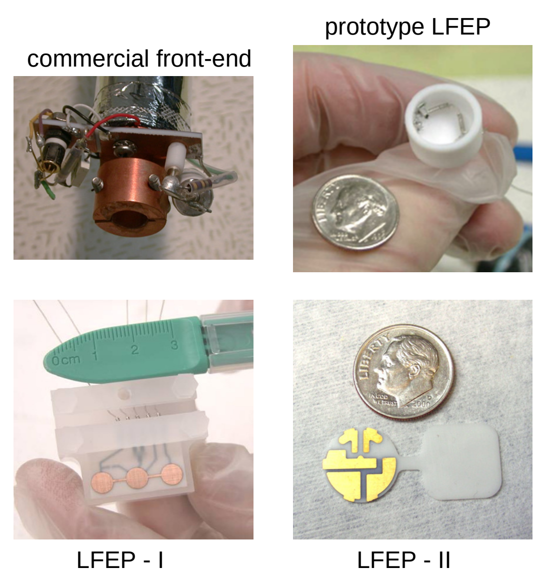

Figure 8. Evolution of front-ends from commercial unit to current LFEP-II.

Production of Final Parts

Final parts for the first cryostat were machined over the summer; assembly and test of the cryostat is underway. Figure 9 shows the cryostat cross-arm and mandrel being turned to final dimensions on a lathe. In order to minimize surface contaminants, the machinists are required to wear gloves while handling the copper parts to improve cleaning results at later stages. Machine tool parts that contact the piece were also cleaned prior to work, and clean propylene glycol is

used to lubricate the surfaces during machining.

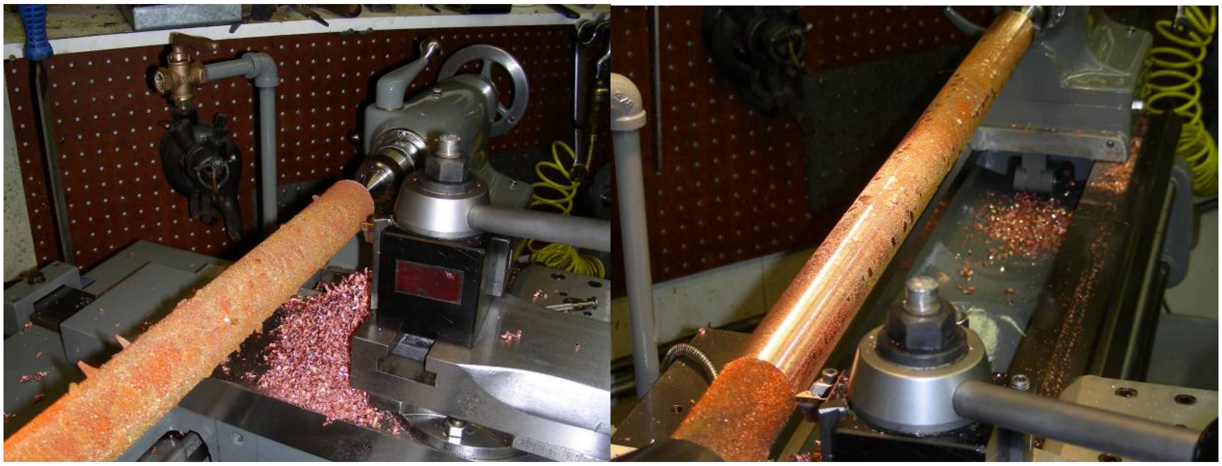

Figure 9. Initial lathe turning of the cold finger for the first 7-crystal cryostat. Shavings are from truing the rod ends and starting to remove the nodule growths from the electroformed piece. Care must be taken during this stage to avoid tearing the nodules away and leaving pits that extend inside the finished diameter.

CONCLUSIONS AND RECOMMENDATIONS

Construction of the first of two ultra-low-background cryostats, each to house 7 HPGe crystals, is underway. Electroforming of parts for the 2nd cryostat is also in progress. Initial post-construction testing will include an assessment of vacuum and thermal performance, however test results are not available at the time of this writing. The crystals will be installed after successful check-out of the cryostat.

Simulation results for the system indicate that the potential to establish detection sensitivity levels in the range of mid 10^4 fissions for samples measured fivedays post-event, with a 24-hour measurement and no chemical processing of the sample.

Achieving the lowest possible backgrounds is an important part of overall system performance. Accordingly, deployment to a shallow underground or deeper operating location (if available), will maximize the sensitivity performance of this system.

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